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SUMMARY
Data on the impact of domestic woodburning on indoor air quality (IAQ) is poor whereas use of such heating systems increase with the development of biofuels. This project aims at characterising IAQ in single family dwellings burning wood.

Field investigations were performed in three occupied houses respectively equipped with an open fireplace, an old and a recent woodstoves. Continuous measurements of air temperature, relative humidity, carbon monoxide, nitrogen oxides, polycyclic aromatic hydrocarbons (PAHs) were performed in the room with the appliance. Moreover in this room and in a bedroom, PM_{10} and $PM_{2.5}$, PAHs (on PM_{10} fraction), volatile organic compounds (VOCs), aldehydes, ketones, and tracers of wood combustion such as levoglucosan and methoxyphenols, were measured. The measurements were operated before and during woodburning; nominal and reduced burning conditions were tested. Permeability and ventilation were assessed using different techniques: blower door, tracer gas, visual inspection, and CO2 or exhausted airflow rate measurements.

KEYWORDS
Woodburning, PAHs, Particulate matter, VOCs

INTRODUCTION
Data on the impact of domestic woodburning appliances on indoor air quality in industrialised countries is rather poor. Even if the International Agency for Research on Cancer (IARC) classified woodsmoke as probably carcinogen for Humans in 2006 (IARC, 2006), less than 10 studies (from US, Canada and Sweden mainly) have been collected by a scientific literature review. It is thus particularly difficult to extrapolate theses results to all types of woodburning situations since appliances, burning conditions, building characteristics, are not always precisely described and are anyway very different. However, according to these few studies, residential woodburning would not have any impact on aldehydes and nitrogen dioxide indoor concentrations, since these compounds are also emitted by numerous other indoor sources. Concerning polycyclic aromatic hydrocarbons (PAHs), benzene and 1,3-butadiene, indoor levels seem to be higher in the presence of a woodburning (open fireplace or woodstove). Finally woodburning impact on particulate matter and carbon monoxide indoor levels have been insufficiently studied to date.

Currently use of such heating systems increase as the promotion of biofuels is developing. According to national inventories, woodburning represents a non-negligible source of organic pollutants in ambient air. Consequently our study aims at describing both emission factors and

indoor and outdoor concentrations of a wide range number of pollutants emitted by different types of woodburning appliances (open fireplaces and woodstoves). Global sampling strategy and first results for indoor environment are presented hereafter.

MATERIALS & METHODS

Global study design

Field investigations were performed in February 2007 in three single family occupied houses in rural areas located North from Paris (70 km). In rural areas woodburning is widely used and it is easier to study the impact on ambient air since other sources like traffic or industries are not predominant.

To fit to the French typical existing equipment, three different appliances were selected. One house was equipped with an open fireplace, one with an old woodstove (about 15 years old) and the last one with a recent woodstove (2004). The houses were chosen among volunteers first contacted through email. Smokers' homes, or houses with chimney not easily accessible on the roof, with attached garages, or with non representative appliance (old type or with particularly high efficiency), were systematically excluded. Standardised wood logs (identical humidity content in particular) were provided by the French National school of wood industries for all the three campaigns.

Houses were normally inhabited during the sampling period: occupant activities have been described through a questionnaire. Cooking took place, but no smoking. Main door opening has been consigned; occupants were asked not to open the other external doors and windows during the sampling periods from 9 am to 5 pm.

Sampling

The selection of compounds to be measured was based on the fact that:

- they are emitted by woodburning (PM_{10}, $PM_{2.5}$, PAHs, dioxins/furans…), and few of them are known to be tracers of this type of combustion such as levoglucosan, and methoxyphenols. They may have already been studied to assess the impact of woodburning on indoor air quality (Strandberg et al., 2006);
- surveillance of compounds in ambient air is mandatory (nitrogen oxides (NOx); sulfur dioxide (SO_2); ozone (O_3), benzo[a]pyrene…) (EC, 1999, 2002, 2004);
- substances are "classical" indoor air pollutants (i.e. formaldehyde, benzene…), and indoor air quality guidelines based on health criteria have already been established by international bodies (INDEX, 2005);
- compounds are of very high concern, as regards health effects, such as carbon monoxide (CO) (WHO, 2000);
- parameters or substances are usually measured to describe ambient conditions (carbon dioxide (CO_2); oxygen (O_2); temperature, relative humidity).

The list of measured parameters and compounds is summarised in Table 1.

Each home was investigated one week long (5 days from Monday to Friday), including 3 days of measurements. During the first one (day 2), measurements were operated (indoor and outdoor) without woodburning. Then both nominal and reduced burning conditions were tested, respectively during day 3 and day 4. Outdoor samplers were placed in two secure locations, close to the house (in the front or back yard) and approximately 200 m far away in the village to assess background ambient air quality. Indoor air was sampled in the living-

room, where the appliance was located, and in the main bedroom (same floor or on the first floor). Exhaust fumes at the chimney on the roof were sampled on day 3 and day 4.

Table 1. List of selected (x) compounds and parameters.

Compounds	**Emission**	**Indoor**		**Outdoor**
		Living-room	**Bedroom**	
CO	x	x	x	
CO_2	x			
O_2	x	x	x	
Total VOCs	x			
VOCs including benzene, toluene, xylenes	x	x	x	x
PM_{10} and $PM_{2.5}$	x	x	x	x
NOx	x	x	x	x
O_3				x
SO_2				x
Aldehydes and ketones (C_1 to C_9)	x	x	x	x
PAHs	x	x	x	x
Levoglucosan	x	x	x	x
Methoxyphenols (guaiacol, syringol for ex.)	x	x	x	x
Dioxins/furans	x			
Temperature	x	x	x	x
Relative humidity	x	x	x	x

Inside the house and in both investigated rooms, PM_{10} and $PM_{2.5}$ were collected on quartz filters for a differed gravimetric analysis. Levoglucosan and methoxyphenols were also sampled on quartz filters in the PM_{10} fraction. Both particulate (quartz filters) and gas (ORBO™ 43 tubes) phases of PAHs (18 congeners) were sampled using a modified ChemPass sampler (R&P) with a PM_{10} head. Canister and Sep-Pack cartridges were respectively used for VOC and aldehyde/ketone sampling. These samplings were all performed 8 hour long in the middle of each room, at about 1.5 m from the ground. Woodburning appliance was in use (nominal or reduced) all along the sampling period.

Inside the house, but only in the living-room, continuous on-line measurements were performed for:

- NOx: chemiluminescent analyser (Thermo Environment Instruments 42C);
- PAHs: photo-ionisation analyser (ECOCHEM PAS-2000) ;
- CO and O_2: multigaz sensor (OLDHAM MX2100);
- temperature: PT100 probe.

Analytical techniques

PAHs

Filters and ORBO tubes were extracted together by sonication with acetonitrile. The extracts were filtered and concentrated under a nitrogen stream (Zymark Turbovap II). 18 PAHs were quantified by HPLC with fluorescence/UV detection.

Aldehydes/ketones
DNPH cartridges were extracted with acetonitrile. 10 aldehydes and 6 ketones were quantified by HPLC/UV detection.

VOCs
Canisters were analysed with a TurboMatrix TD-GC system equipped with an on-line sampling accessory and two FID detectors. The GC contained two columns in order to analyse VOCs from C_2 to C_{11}. 34 VOCs were quantified.

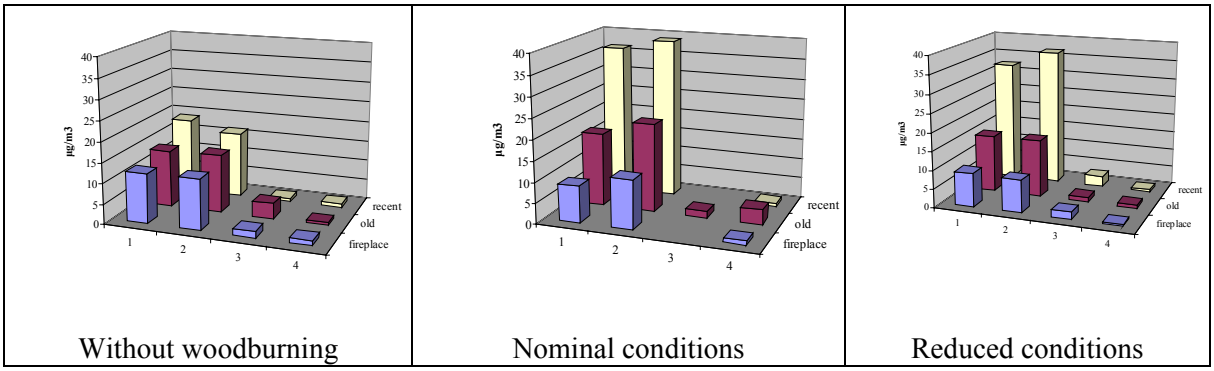
Permeability and ventilation assessment
Since ventilation and permeability measurements may be quite long and difficult to operate in all the investigated homes, different quantitative or qualitative strategies were adopted.

In only one dwelling, air leakage of the building envelope was assessed using blower door technique. The home is artificially placed in depression thanks to a fan on the door generating an outdoor-indoor pressure difference. Extracted air flow is then measured with diaphragms. Such measurements can easily be interfered and for example should not be operated if the wind speed is above 2 m/s. Moreover in this house the air renewal rate was measured in the living-room and the bedroom using tracer gas technique by the decay method. SF6 was the reference gas: detection limit 5 ppm, linearity up to 500 ppm between 5 and 40°C, 8% uncertainty factor. During the daily heating and sampling period (8 hours), 3 decays could be measured in each operated room.

In the two others dwellings, magnitude of air exchange was qualitatively assessed through visual inspection of the envelope components: date of construction, eventual rehabilitation, orientation regarding dominant winds, room and stage numbers, construction materials, type of isolation, type of windows, natural or mechanical ventilation... In case of mechanical ventilation, exhausted airflow rate was measured at the air outlets with a SWEMAFLOW 233 flow meter. Finally spot measurements of CO2 were also used to assess air exchange rate.

RESULTS
First indoor measurement results are presented on Figure 1 to Figure 4. Organic tracers (levoglucosan, mannosan, methoxyphenols) were not detected indoors. This could be due to too low sampling volume limiting the quantity of collected tracer to a value lower than the detection limit; for the future measurements a higher air volume will be sampled.

Caption for all the Figures:
1: indoor, living-room; 2: indoor, bedroom; 3: outdoor, close to the house; 4: outdoor, background
recent = recent woodstove; old = old woodstove; fireplace = open fireplace



Without woodburning | Nominal conditions | Reduced conditions

Figure 1. Formaldehyde concentrations (µg.m-3).

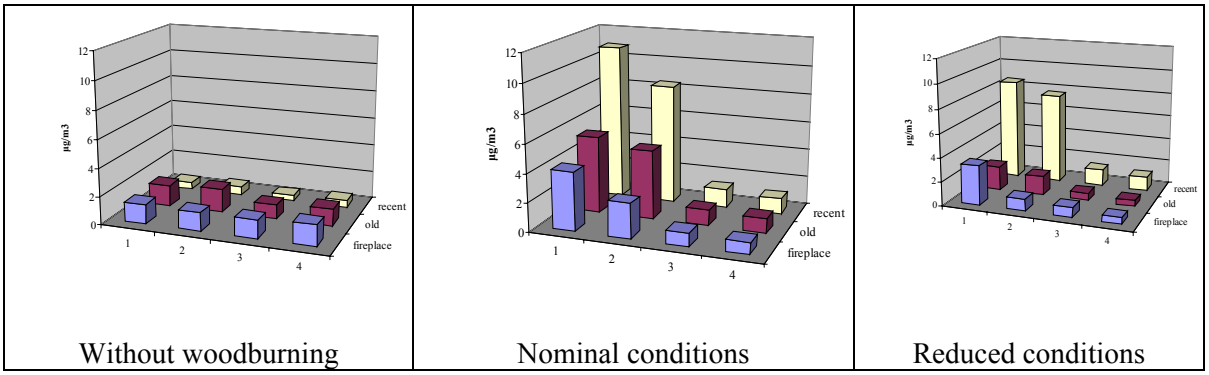


Figure 2. Benzene concentrations (µg.m-3).

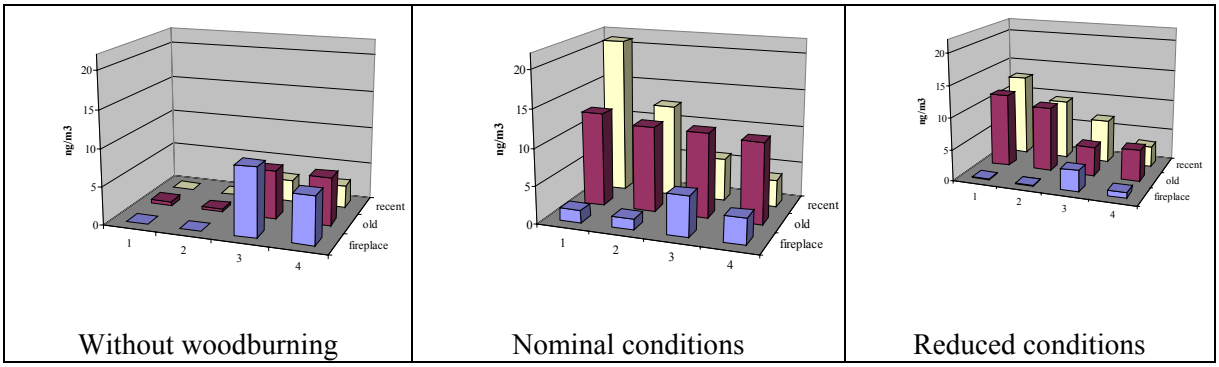


Figure 3. Particulate PAH concentrations (ng.m-3).

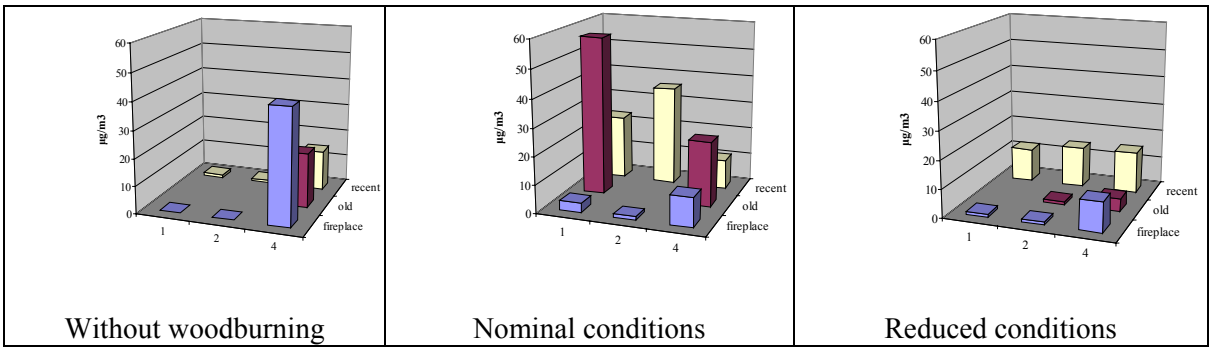


Figure 4. $PM_{2.5}$ concentrations (µg.m-3).

DISCUSSION

Data analysis is still undergoing. Nevertheless we observe that:

- woodburning seems having minor impact on indoor air quality regarding formaldehyde. Similar concentrations have been measured in the kitchen and the bedroom on the days with wood appliance under heating. Meanwhile benzene, particulate PAHs and $PM_{2.5}$ levels seem to be significantly influenced by woodburning, in particular by woodstoves, even in the room distant to the appliance (the bedroom in our study);
- formaldehyde and benzene indoor levels show no noticeable difference between nominal and reduced burning conditions. However reduced conditions would have a worsen impact on particulate PAHs indoor concentrations;
- comparing the three appliances, the fireplace appears to have much less impact on indoor air quality than the two woodstoves. Age of these latter does not appear to be a significant parameter for the appliances which were tested. No reason can be found to explain that the old woodstove has less impact than the more recent one.

These first conclusions have to be analysed regarding ventilation (e.g. air exchange rate) and building permeability.

Indoor concentrations are compared to levels measured in French dwellings by the French Indoor Air Quality Observatory (OQAI) between 2003 and 2005 during its national "Housing campaign" (OQAI, 2006). The 567 investigated homes were representative of the 24 million French metropolitan main residences. Benzene and formaldehyde were sampled one week long (7 days) in the bedroom through passive diffusive samplers. $PM_{2.5}$ were measured in the living-room in the presence of occupants (from 5 pm to 8 am on weekdays and 24h/24 during the week-end). The indoor geometric means were respectively 2.1 µg.m-3 (percentile95: 7.2 µg.m-3 and maximum value: 22.8 µg.m-3), 19.6 µg.m-3 (percentile95: 46.7 µg.m-3 and maximum value: 86.3 µg.m-3), and 19.1 µg.m-3 (percentile95: 133 µg.m-3 and maximum value: 368 µg.m-3). In comparison indoor benzene levels while woodburning did not exceed the maximum value, but were globally higher than the geometric mean value. It should be noted that before burning wood, indoor benzene concentrations were below this mean value in all the three homes. Formaldehyde indoor levels were in the same order of magnitude or even lower than the geometric mean, before or during woodburning, except for the case of the recent stove for which indoor levels are higher but in the range of previously encountered indoor levels. Finally $PM_{2.5}$ indoor concentration exceed the geometric mean value in one situation (old stove, nominal conditions, sampler in the kitchen), but remain far below the maximum value observed by the French IAQ Observatory.

Due to the lack of a French reference PAHs set of data, levels were compared with those from the American RIOPA (Relationships of Indoor, Outdoor, and Personal Air) study (Turpin et al, 2007). PAHs on $PM_{2.5}$ fraction were analysed in approximately 100 homes within each of three American cities (Los Angeles, Elizabeth, and Houston). Even if the indoor levels are not rigorously comparable because of different sampled and analysed granulometric fractions, it is clearly observed that indoor particulate PAHs measured in our study were much higher than the RIOPA mean values, for all the PAHs congeners (up to more than 2 orders of magnitude).

CONCLUSIONS

Even results should be completed and refined, we can however conclude that we observe an impact of woodburning with stoves on indoor air quality within the three single family houses investigated in February 2007. This impact seems globally to depend preferentially on the type of appliance rather than on burning conditions (nominal or reduced). The magnitude of this impact is various depending on the compounds that are considered; major influence of woodburning is observed on benzene and particulate PAH indoor levels.

Another set of measurements lead in November 2007 in three other houses in rural areas following the same protocol, is currently under analysis; this additional set of values will be of high interest to confirm our preliminary observations.

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REFERENCES

EC. 1999. Official Journal of The European Communities. Directive 1999/30/CE of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air. DOCE L163, 29/6/1999.

EC. 2002. Official Journal of The European Communities. Directive 2002/3/EC of 12 February 2002 relating to ozone in ambient air. DOCE L67/14, 9/03/2002.

EC. 2004. Official Journal of The European Communities. Directive 2004/107/CE of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air. OJ L 23, 26.1.2005, pp. 3–16.

IARC. 2006. Indoor emissions from household coal combustion carcinogenic: women in low- or medium-resource countries most exposed, International Agency for Research on Cancer, Press Release N° 172.

INDEX. 2005. European Commission. Joint Research Centre (JRC). Final Report. Critical Appraisal of the Setting and Implementation of Indoor Exposure Limits in the EU : The INDEX project. Institute for Health and Consumer Protection, Physical and Chemical Exposure Unit. 337 pages. I-21020 Ispra (VA), Italy.

OQAI. 2006. Observatoire de la Qualité de l'Air Intérieur, Campagne nationale Logements. Etat de la qualité de l'air dans les logements français. Final report, 165 pages, in French.

Strandberg B., Gustafson P., Söderström H. et al. 2006. The use of semipermeable membrane devices as passive samplers to determine persistent organic compounds in indoor air. Journal of Environmental Monitoring 2006, 8: 257-62.

Turpin B.J., Weisel C.P., Morandi M. et al. 2007. Relationships of Indoor, Outdoor, and Personal Air (RIOPA). Part II, Analyses of Concentrations of Particulate Matter Species. Health Effects Institute. 110 pages.

WHO. 2000. WHO Air Quality Guidelines for Europe, second edition No.91.